INFRARED ABSORPTION BY SHAPE DISTRIBUTIONS OF NH_3 ICE PARTICLES: AN APPLICATION TO THE JOVIAN ATMOSPHERE

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Abstract. We study how distribution of small NH_3 ice particles over shapes affects the strength of resonant absorption features at 9.4 and 26 μm. The *T*-matrix approach is used to compute optical cross sections for shape distributions of 0.5- and 1-μm volume-equivalent radius spheroids in random orientation. It is found that the maximum of the resonant absorption for the shape distributions is 1.5–2 times smaller than that for equal-volume spherical particles, the absorption peak being shifted towards longer wavelengths. The results of our computations support the conclusion of West *et al.* (1989) that, apparently, small NH_3 ice particles cannot be the principal component of the Jovian troposphere in the 300- to 500-mbar region.

1. Introduction

It is usually assumed that the upper tropospheric aerosols in the Jovian atmosphere mainly consist of ammonia ice (see, e.g., the review by West *et al.*, 1986). There is an evidence that the top of the upper tropospheric cloud is in the 200- to 300-mbar region, and that the aerosols in the 300- to 500-mbar region (or at optical depths up to several units) have sizes near 1 μm or smaller (e.g., Morozhenko and Yanovitskii, 1973; West *et al.*, 1985; West *et al.*, 1986; Mishchenko, 1990b). As it follows from the Mie theory, the absorption spectra of submicron-sized spherical NH_3 ice particles have a strong absorption feature near 9.4 μm and a broader but recognizable feature near 26 μm. These absorption features are due to the surface resonance modes which occur at that wavelengths where the real part of the refractive index is close to zero, and the imaginary part is near $\sqrt{2}$ (see, e.g., Chapter 12 of Bohren and Huffman, 1983). Thus, small spherical NH_3 ice particles in the upper Jovian troposphere should produce easily detectable absorption features in the Jovian spectrum near 9.4 and 26 μm. Nevertheless, these absorption features are not seen in the observed spectra of Jupiter, which leads to the conclusion that (i) the aerosols in the upper Jovian troposphere are small but do not consist of NH_3 ice, or (ii) these aerosols consist of NH_3 ice but are not small, or (iii) these aerosols are small and consist mainly of NH_3 ice, but the resonance absorption features are washed out due to some causes (see West *et al.*, 1986 and West *et al.*, 1989 for discussion and references).

It is well known that the following three factors can significantly influence the strength and shape of the infrared resonance absorption features: (1) the inclusion

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of foreign material into the scattering ice particles, (2) nonsphericity of the particles, and (3) distribution of the particles over shapes (see, e.g., Bohren and Huffman, 1983). The first two factors were recently analysed by West *et al.* (1989). They have computed extinction cross-sections for 0.5- and 1-μm volume-equivalent radius tetrahedral NH_3 ice particles with and without nonabsorbing foreign cores. It was found that the small single-shaped homogeneous particles composed of NH_3 ice cannot explain the absence of the absorption features in the Jovian spectrum near 9.4 and 26 μm, and that a large fraction of the particles must be something other than NH_3 ice if the absorption features are to be suppressed by foreign inclusions.

The purpose of the present paper is to continue the work by West *et al.* (1989) by analysing how the strength of the resonance absorption features is affected by distribution of small NH_3 ice particles over shapes. As is demonstrated by terrestrial H_2O ice clouds (see, e.g., Mazin and Khrgian, 1989), a great variety of particle shapes can be expected in crystal clouds. Nevertheless, in this paper we consider only the simplest nonspherical particles – namely, oblate and prolate spheroids – our reasons being as follows. Firstly, light scattering computations for arbitrarily shaped nonspherical particles are very complicated, and simple particle shapes should be chosen to make the problem solvable. Secondly, although real NH_3 ice particles are not more spheroidal than they are spherical or tetrahedral, we hope that by averaging over a representative sample of spheroidal shapes we can obtain rather reliable quantitative estimates of the effect considered; this representative sample of spheroidal shapes, ranging from "needles" to "plates", can be obtained by varying only one parameter: namely, the ratio of the semi-axes of the spheroid.

2. Computational Method

In our computations, Waterman's (1971) *T*-matrix approach was used (see also Barber and Yeh, 1975; Ström, 1975; Tsang *et al.*, 1984). By use of this approach the fields, which are incident upon (superscript *i*) and scattered by (superscript *s*) a single nonspherical particle in a fixed orientation, are expanded in the vector spherical wave functions as (cf. Tsang *et al.*, 1984)

$$\mathbf{E}^i(\mathbf{r}) = \sum_{mn} [a_{mn} \mathrm{Rg} \mathbf{M}_{mn}(\mathbf{kr}) + \mathrm{b}_{mn} \mathrm{Rg}\, \mathbf{N}_{mn}(k\mathbf{r})] \tag{1}$$

and

$$\mathbf{E}^s(\mathbf{r}) = \sum_{mn} [p_{mn} \mathbf{M}_{mn}(k\mathbf{r}) + q_{mn} \mathbf{N}_{mn}(k\mathbf{r})], \tag{2}$$

where $k = 2\pi/\lambda$, and λ is a free space wavelength. The functions Rg $\mathbf{M}_{mn}$ and Rg $\mathbf{N}_{mn}$ in Equation (1) have a Bessel function radial dependence, while the functions $\mathbf{M}_{mn}$ and $\mathbf{N}_{mn}$ in Equation (2) have a Hankel function radial dependence. The expansion coefficients of the incident field a_{mn} and b_{mn} are assumed to be known (e.g., for a plane incident wave they are given by simple analytical expressions), whereas the expansion coefficients of the scattered field p_{mn} and q_{mn} are initially unknown. The relation between these coefficients is linear and is given by a transition matrix (or T-matrix) $\mathbf{T}$ as

$$\begin{bmatrix} \mathbf{p} \\ \mathbf{q} \end{bmatrix} = \mathbf{T} \begin{bmatrix} \mathbf{a} \\ \mathbf{b} \end{bmatrix} = \begin{bmatrix} \mathbf{T}^{11} & \mathbf{T}^{12} \\ \mathbf{T}^{21} & \mathbf{T}^{22} \end{bmatrix} \begin{bmatrix} \mathbf{a} \\ \mathbf{b} \end{bmatrix}, \tag{3}$$

where compact matrix notations are used. The elements of the T-matrix do not depend upon the directions of propagation and states of polarization of the incident and scattered fields. They depend only upon the size, morphology, and composition of the scattering particle, as well as upon its orientation with respect to the coordinate system chosen. The T-matrix can be computed by using special formulae, and then one easily computes the expansion coefficients of the scattered field and the scattered field itself (see Equations (3) and (2), respectively), as well as the amplitude scattering matrix, optical cross sections, single scattering albedo, etc.

The T-matrix approach is especially efficient for axially symmetric scattering particles, since in this case the T-matrix can be diagonalized with respect to the indices m and m' by using the natural (or body) coordinate system with the z-axis along the axis of particle symmetry (see, e.g., Tsang *et al.*, 1984): i.e.,

$$\hat{T}^{ij}_{mnm'n'} = \delta_{mm'} \hat{T}^{ij}_{mnmn'}, \quad i, j = 1, 2, \tag{4}$$

where $\delta_{mm'}$ is the Kronecker delta, and $\hat{\mathbf{T}}$ is the T-matrix of the axially symmetric particle computed in the natural coordinate system. Formulae for computing the matrix $\hat{\mathbf{T}}$ are given, e.g., by Tsang *et al.* (1984). Numerical aspects of the T-matrix computations are considered in detail by Wiscombe and Mugnai (1986).

An important advantage of the T-matrix approach is that it is ideally suited to calculate orientationally averaged quantities for randomly oriented or partially aligned nonspherical particles (Varadan, 1980; Tsang *et al.*, 1984; Mishchenko, 1990a, c–f). In particular, we have shown (Mishchenko, 1990a, d) that the extinction and scattering cross sections, averaged over the uniform orientational distribution of identical axially symmetric particles, are given, respectively, by

$$C_{\mathrm{ext}} = -\frac{2\pi}{k^2} \operatorname{Re} \sum_{n=1}^{\infty} \sum_{m=0}^{n} (2 - \delta_{m0}) (\hat{T}^{11}_{mnmn} + \hat{T}^{22}_{mnmn}) \tag{5}$$

and

$$C_{\mathrm{sca}} = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \sum_{m=0}^{\min(n,n')} \sum_{i,j=1,2} (2 - \delta_{m0}) |\hat{T}^{ij}_{mnmn'}|^2. \tag{6}$$

3. Numerical Results

As it was pointed out in Section 1, to compute the resonance absorption features for shape distributions of nonspherical NH_3 ice particles, we used mixtures of prolate and oblate spheroids of various aspect ratios. The shape of a spheroid in its natural coordinate system is governed by the equation

$$r(\theta, \varphi) = a\left(\sin^2\theta + \frac{a^2}{b^2}\cos^2\theta\right)^{-1/2}, \tag{7}$$

where b is the rotational semi-axis, and a is the horizontal semi-axis of the spheroid. Another pair of parameters, that may be used to specify the shape of the spheroid, is (r_{ev}, d), where $d = a/b$ is the ratio of the semi-axes, and r_{ev} is the radius of the equal-volume sphere given by

$$r_{\mathrm{ev}} = ad^{-1/3}. \tag{8}$$

In order to make the mixture of particle shapes representative enough, on the one hand, and to avoid numerical instability of the T-matrix computations, on the other, we used the values of the parameter d from the interval [1/8, 8]. We included in the mixture fifteen equal-volume spheroids with d = 1/8, 1/7, 1/6, 1/5, 1/4, 1/3, 1/2, 1, 2, 3, 4, 5, 6, 7, and 8. For the sake of simplicity, all the spheroidal shapes were assumed equally probable. Thus, the ensemble averaged extinction and scattering cross sections were computed as

$$\langle C_{\mathrm{ext}}\rangle = (1/15)\sum_{n=1}^{15} C^n_{\mathrm{ext}} \tag{9}$$

and

$$\langle C_{\mathrm{sca}}\rangle = (1/15)\sum_{n=1}^{15} C^n_{\mathrm{sca}}, \tag{10}$$

where n numbers the spheroidal shapes. Then, the single scattering albedo w was computed as

$$w = \langle C_{\mathrm{sca}}\rangle/\langle C_{\mathrm{ext}}\rangle. \tag{11}$$

The computational parameters, that determine the accuracy of the T-matrix computations (see, e.g., Wiscombe and Mugnai, 1986), were set such that the accuracy of computing the ensemble averaged cross-sections was better than 0.3%. Refractive indices of NH_3 ice were taken from Martonchik *et al.* (1984).

Computational results for two mixtures of randomly oriented spheroids with $r_{\mathrm{ev}} = 0.5$ and 1 μm are shown in Figures 1–4 and Table I. For comparison, analogous computations for equal-volume spherical particles are also displayed. Instead of the extinction cross-sections, efficiency factors for extinction are plotted, which are defined as

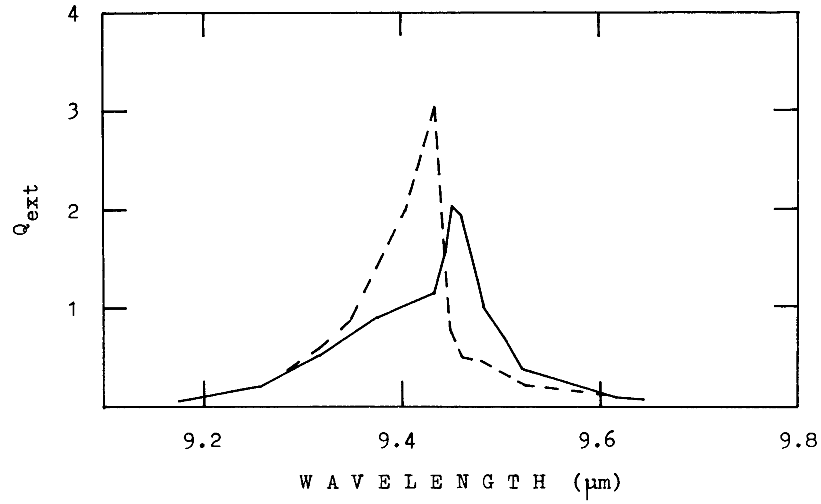

Fig. 1. Extinction efficiency factors for the mixture of spheroids with $r_{ev} = 0.5\ \mu m$ (solid line) and the equal-volume sphere (dashed line).

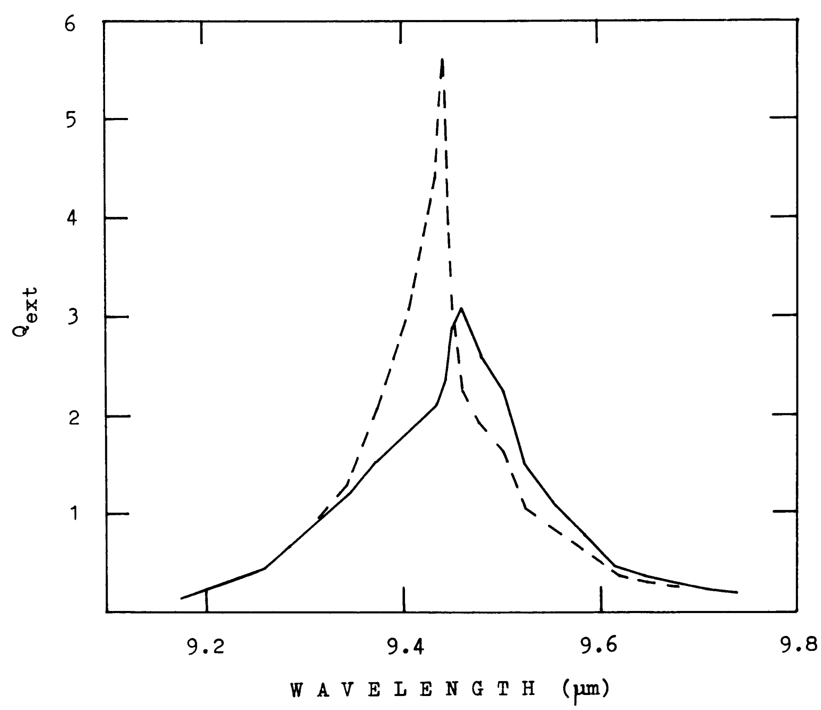

Fig. 2. As in Fig. 1, for $r_{ev} = 1\ \mu m$.

$$Q_{ext} = \langle C_{ext} \rangle / S, \tag{12}$$

where $S = \pi r_{ev}^2$ is the geometrical cross-section of the equal-volume sphere.

It is interesting to note that our curves for the shape mixtures of spheroidal particles look very much like those computed by West *et al.* (1989) for equal-volume tetrahedra. One sees that the maximum of the resonance absorption for

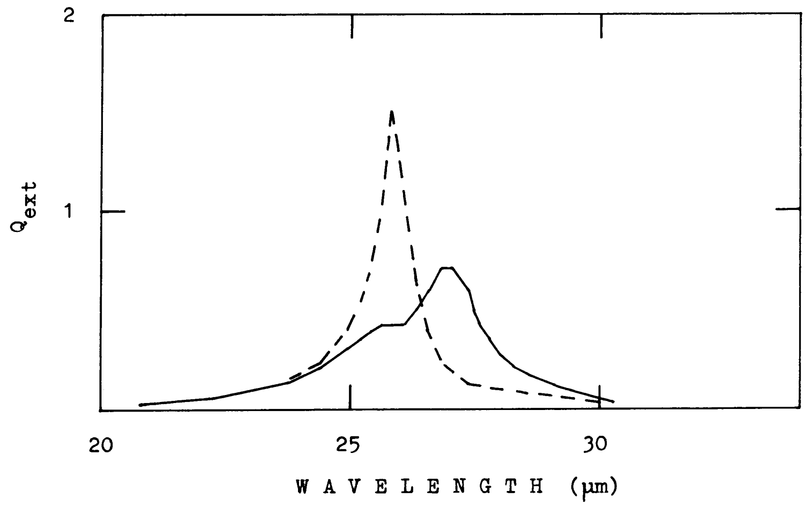

Fig. 3. As in Fig. 1, for the 26 μm region. In the 20- to 30-μm region, the single scattering albedo is less than 0.01.

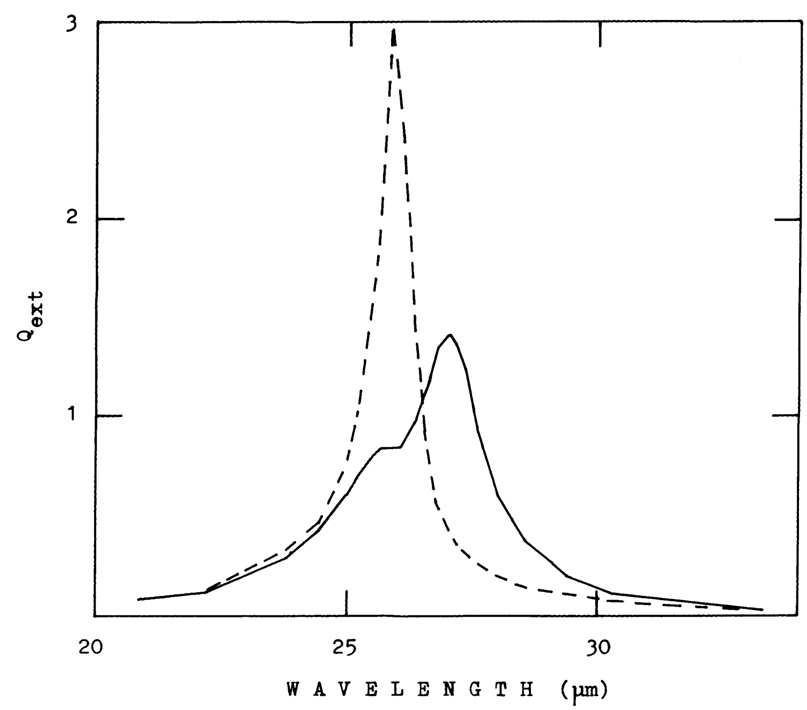

Fig. 4. As in Fig. 2, for the 26 μm region. In the 20- to 30-μm region, the single scattering albedo is less than 0.04.

the shape distributions is 1.5–2 times smaller than that for the equal-volume spherical particles, the absorption peak being shifted towards longer wavelengths. Nevertheless, these changes seem to be not sufficient to explain the fact that the absorption features are not seen in the Jovian spectra.

TABLE I

Spectral values of the single scattering albedo for the 9.4-μm region

λ, μm	$r_{ev} = 0.5\ \mu$m		$r_{ev} = 1\ \mu$m	
	Spheroids	Sphere	Spheroids	Sphere
9.01	0.00	0.00	0.02	0.02
9.20	0.01	0.01	0.04	0.05
9.29	0.01	0.02	0.09	0.10
9.37	0.03	0.03	0.15	0.17
9.44	0.11	0.11	0.41	0.47
9.48	0.08	0.07	0.32	0.32
9.55	0.08	0.08	0.34	0.33
9.65	0.13	0.13	0.48	0.49
9.74	0.16	0.17	0.56	0.58

4. Conclusions

In the present paper we analysed how distribution of small NH_3 ice particles over shapes can affect the strength of the resonance absorption features near 9.4 and 26 μm. Our work is a continuation of the paper by West *et al.* (1989), where two other factors were analysed: namely, nonsphericity of NH_3 ice particles of a single (tetrahedral) shape and inclusion of foreign material into the ice particles. Both our computations and those of West *et al.* (1989) show that cloud particles of sizes near 1 μm or smaller in the upper Jovian troposphere should produce easily detectable absorption features near 9.4 and 26 μm in the Jovian spectrum if they mainly consist of NH_3 ice. Thus, following West *et al.* (1989), we conclude that, apparently, small NH_3 ice particles cannot be the principal component of the upper Jovian troposphere.

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